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(54) Fuel cell membrane electrode and seal assembly.

(57) A fully supported membrane assembly (40) for an electrochemical fuel cell is provided. A first and second layer of porous electrically conductive sheet material (44,50), such as carbon fiber paper, has a solid polymer ion exchange membrane (43) interposed therebetween. The layers of sheet material cover and support the membrane over substantially its entire surface area. The layers of sheet material are coated with a catalyst (54) to tender them electrochemically active. The layers of sheet material and the membrane are bonded together to form a consolidated assembly. Openings (112,111,113) are formed in the layers of sheet material and the membrane to accommodate the passage of fluids through the assembly. Seals are formed by impregnating the layers of sheet material with a sealant material which generally circumscribes the fluid passage openings and the electrochemically active portion of the assembly. Alternatively, grooves are formed in the surfaces of the electrodes facing away from the membrane, and sealant material is deposited into the grooves. The grooves generally circumscribe the fluid passage openings and the electrochemically active portion of the assembly.

Fig. 5

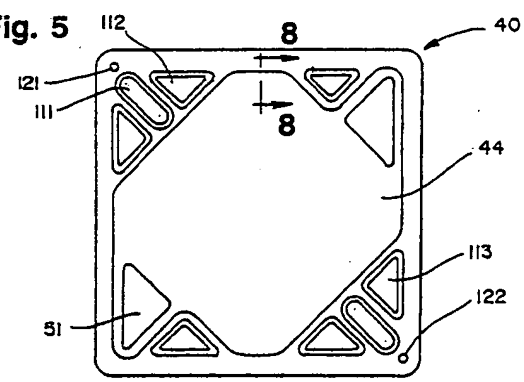
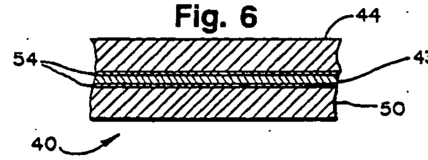


Fig. 6



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also necessary to machine a recess in each flow field plate contiguous with the periphery of the CFP so that the MEA could be appropriately positioned between the flow field plates with a uniform distance being maintained about the periphery so that the membrane could be tightened appropriately between the plates and function with a good sealing action. Such machining was time consuming and expensive and, in fact, did not assist substantially in enhancing the sealing action.

Yet a further disadvantage with the conventional membrane electrode assembly was that the membrane itself was difficult to position and maintain in position while the stack assembly was being assembled. This was so since the membrane is quite thin and is inherently very flexible. It was also being subject to expanding and contracting due to the humidity changes in the gases to which the membrane was subjected.

Yet a further disadvantage in the prior membrane electrode assembly was the problem of positioning seals about the water and gas passages which extended through the flow field plates and the membrane. This was accomplished by machining grooves in the graphite flow field plates on either side of the membrane assembly and manually positioning rubber seals in the grooves. This was time consuming and, when assembling the cells, the seals could be dislodged from the grooves if of an O-ring configuration. If the seals took a rectangular configuration, they could be rolled in their grooves. In either case, the sealing action was adversely affected.

SUMMARY OF THE INVENTION

According to the invention, a membrane assembly for an electrochemical fuel cell comprises first and second layers of porous electrically conductive sheet material and a membrane interposed therebetween. The layers of sheet material cover and support substantially the entire surface of the membrane.

In the preferred embodiment, the membrane is a solid polymer ion exchange membrane and the sheet material comprises carbon fiber paper. The membrane most preferably has a thickness between about .001 inches and about .005 inches.

In the membrane assembly of the active section of the fuel cell, each of the layers of sheet material further includes a catalyst on at least a portion thereof. The layers of sheet material are thereby rendered electrochemically active in the area containing the catalyst. The preferred catalyst comprises platinum. The catalyst is preferably included on at least a portion of the surface of each of the layers of sheet material facing the membrane. Most preferably, the catalyst is included on the central portion of each of the layers of sheet material. The layers of sheet material and the membrane are preferably bonded together to form a consolidated membrane electrode assembly.

In the preferred membrane electrode assembly, the layers of sheet material and the membrane have openings formed therein to accommodate the passage of fluids through the assembly. The fluids include fuel gas, oxidant gas and coolant. The preferred coolants are water and ethylene glycol.

In one embodiment of the membrane electrode and seal assembly, each of the layers of sheet material has grooves formed in the surface thereof facing away from the membrane. The grooves have an extrudable sealant material deposited therein, and generally circumscribe the fluid passage openings. A groove generally circumscribing the electrochemically active portion of the assembly can also be formed in each of the layers of sheet material. The sealant material preferably extends within the pores of the sheet material and protrudes above the surfaces facing away from the membrane. The grooves preferably extend substantially the entire thickness of each of the layers of sheet material, and in the most preferred embodiment, the sealant material contacts the membrane. The sealant material preferably comprises silicon rubber.

In another embodiment of the membrane electrode and seal assembly, the surfaces of the layers of sheet material facing away from the membrane are impregnated with a sealant material. The sealant material generally circumscribes the fluid passage openings. The sealant material can also circumscribe the electrochemically active portion of the assembly. The sealant material preferably protrudes above the surfaces facing away from the membrane, and extends substantially the entire thickness of the layers of sheet material. In the most preferred embodiment, the sealant material contacts the membrane. The sealant material preferably comprises silicon rubber.

A first method of forming a membrane assembly comprises the steps of:

bonding a solid polymer ion exchange membrane between two layers of porous electrically conductive sheet material, the layers covering and supporting substantially the entire surface of the membrane, the layers and the membrane having openings formed therein to accommodate the passage of fluids through the assembly,

forming grooves in the surfaces of the layers facing away from the membrane, the grooves generally circumscribing the fluid passage openings, and

depositing an extrudable sealant material into the grooves.

along line 8-8 of Figure 5.

DETAILED DESCRIPTION OF THE DRAWINGS

Referring now to the drawings and, in particular, to Figure 1, a fuel cell stack assembly is generally illustrated in exploded form at 10. The stack assembly includes a pair of end plates 11, 12 which conveniently are fluid end plates which terminate the stack assembly 10 and a plurality of threaded tie rods 15 extending between the end plates 11, 12 to retain and hold the stack assembly 10 in its assembled condition.

A spacer plate 13 and an electrical isolation plate 14 are positioned inside the end plate 11 and a piston 17 is positioned within the end plate 12. Buss plates 20, 21 are located on opposite ends of the stack assembly 10 as indicated and carry the voltage and current generated by the fuel cell 10. Cooling water jackets 22, 23 are located immediately inside the buss plates 20, 21.

The stack assembly 10 includes a so-called "active" section generally illustrated at 24 and a "humidification" section generally illustrated at 30. The active section 24 includes in addition to the buss plates 20, 21 and cooling water jackets 22, 23, a plurality of identical assemblies illustrated generally at 31, each assembly consisting of three fluid flow field or graphite plates 32, 33, 34 and two membrane electrode assemblies generally illustrated at 40 which are assembled between the plates 32, 33, 34. In each assembly 31, the leftmost flow plate 32 carries the fuel in the form of hydrogen gas, the center flow plate 33 carries the oxidant in the form of oxygen or air on one side and hydrogen on the opposite side and the rightmost plate 34 carries the oxidant on the side adjacent the assembly 40 and water on the opposite side. The configuration of the assembly 31 thus provides for the hydrogen and the oxidant to be located on opposite sides of each membrane electrode assembly 40 and a coolant fluid flow plate in the form of a coolant jacket which is, in the present instance, a water jacket, to be located adjacent each hydrogen flow field plate. This configuration is typical and extends throughout the active section 24.

The humidification section 30 includes a plurality of oxidant flow field plates 41 generally located on the left hand side of the humidification section 30 illustrated in Figure 1. The humidification section 30 also includes a plurality of fuel humidification flow field plates 42 generally located on the right hand side of the humidification section 30, a plurality of fuel humidification membranes 37 and a plurality of oxidant humidification membranes 36 positioned between the fuel humidification flow field plates 42 and the oxidant flow field plates 41, respectively. The humidification section 30 acts to humidify the gases used so that the solid polymer ion exchange membranes in the active section 24 will remain moist or wet as described hereafter. In general, hydrogen ions will diffuse less readily through solid polymer ion exchange membranes if the membranes are allowed to dry. A lower rate of hydrogen diffusion will in turn reduce overall cell efficiency. The humidification section 30 is intended to prevent the membranes from drying by humidifying the fuel and oxidant gases fed to the active section 24.

The active section 24 also differs from the humidification section 30 in that there is no electricity produced in the humidification section 30 whereas in the active section 24 electricity is produced by reason that a catalyst in the form of platinum is present in the carbon fiber paper forming the membrane electrode assemblies 40 and thus renders a portion of each layer of carbon fiber paper electrochemically active.

Figure 2 is an end view of the fuel cell stack assembly 10 illustrated in Figure 1, showing hydrogen (fuel) gas inlet 71, hydrogen (fuel) gas outlet 72, air (oxidant) inlet 73, air (oxidant) outlet 74, water inlet 75 and water outlet 76. Figure 2 also shows end plate 12, piston 17, and tie rod nuts 120.

The membrane electrode assemblies 40 in the active section 24 are identical and a typical assembly is illustrated at 40 in Figures 5, 6 and 7. Each membrane electrode assembly 40 comprises three elements, namely, a first layer of a porous electrically conductive support material, conveniently a porous carbon fiber paper 44, a second layer of an electrolyte material which is a solid polymer ion exchange membrane 43, and a third layer of a porous electrically conductive support material 50, conveniently formed of carbon fiber paper. The layers of carbon fiber paper 44, 50 support the membrane 43 therebetween to form a consolidated assembly 40 as described in greater detail hereafter. The carbon fiber papers layers 44, 50 are each treated with a catalyst on the surfaces adjacent and in contact with the membrane 43, thus to form electrodes. The treated area coincides with the flow field of the graphite plates which carry the gases to the carbon fiber paper.

A solid polymer ion exchange membrane 43 is conveniently used which has a conventional thickness of approximately .007 inches. This thickness, however, was necessary to reduce tearing and other damage when in an unsupported configuration. It is believed that a reduced thickness will be possible to use with the membrane as supported according to the present invention and, indeed, performance comparisons made to date indicate that with a constant cell terminal voltage, a membrane having a reduced thickness will give en-

The fuel cell 10 is then assembled by using the guide pin holes 121, 122 as guides for guide pins (not shown). The various components are stacked together and, when assembled, the nuts 120 (see Figure 1) on the ends of the tie rods of fuel cell 10 are torqued to apply suitable compressive force.

In operation, the fuel, preferably in the form of hydrogen, flows from the hydrogen supply and enters the humidification section 30 through the fuel humidification flow field plates 42 (see Figure 1) where the fuel gas is humidified by the water carried by the humidified water jacket plates 39 to a value preferably close to 100% relative humidity. The humidified fuel gas then passes to the active section 24 of the stack assembly 10 where the humidified fuel gas passes through the hydrogen or fuel flow field plates 32 adjacent the anode side of the membrane electrode assemblies 40. The oxidant gas supplied to the second electrode and the fuel are consumed by the electrochemical process and an electrical current is generated which is available to be drawn from the porous electrodes. The unused hydrogen exits the plate 32 in the active section 24 and passes to a tank (not shown) where excess hydrogen may be returned to the inlet line 63 and wherein the above process is repeated.

The oxidant in the form of air or oxygen enters the humidification section 30 of the stack assembly 10 where it is humidified as it passes through the oxidant humidification flow field plates 41 (see Figure 1) adjacent the humidification water jacket plates 39. The humidified oxidant gas then passes to the active section 24 and through the oxidant or fuel flow field plates 33, 34 adjacent the cathode side of the membrane electrode assemblies 40. The air or oxygen then flows out of the active section 24 to a separate tank (not shown) where any pressure exceeding a predetermined value is vented and where any water formed is collected for return to the fuel cell 10 for cooling.

In operation, liquid water is provided to the active section 24 of the fuel cell 10 and circulated therethrough to neutralize the heat generated by the exothermic reaction in the fuel cell. The water travels through the water jacket flow field plates 22, 34 adjacent the hydrogen and oxygen flow field plates 32, 33 (see Figure 1). The water then travels to the humidification section 30 and then to a tank where it can be pumped back to the fuel cell 10.

Many modifications will readily occur to those skilled in the art. For example, rather than a graphite material being used for the electrical conducting plates, other substances could be used including a composite material of KYNAR and graphite powder. Likewise, an elastomeric material could be used for the sealant material rather than rubber. It will also be understood that the electrode material may not necessarily be hydrophobic although in the present case, it is so desirable. Although platinum is used as a catalyst in the present instance, it would be possible to use other catalytic substances to promote the reaction. Likewise, while water has been described as being used as coolant, any other suitable coolant, such as ethylene glycol, may be used.

It is further contemplated that rather than machining or milling grooves into the carbon fiber paper which will eventually be used as electrodes and injecting the sealant material into the grooves, grooves need not be formed at all. In this regard, the sealant material could be injected directly into the porous carbon fiber paper to a depth that would contact the membrane without forming grooves and that the quantity of the sealant injected would be such that it would continue to protrude slightly above the surface of the electrode and thereby continue to perform its sealing action in the same manner as set out earlier.

In addition to employing the fully supported membrane electrode and seal assembly in the electrochemically active portion of a fuel cell stack, a fully supported membrane and seal assembly can also be employed in the humidification portion of a fuel cell stack. For ease of manufacture, the membrane assembly in the humidification portion is usually formed of the same membrane and conductive sheet material (preferably carbon fiber paper) as those used in the electrochemically active portion. In the humidification portion, however, the carbon fiber paper is generally not coated with catalyst. As with the membrane assembly used in the electrochemically active portion, it has been found advantageous to employ layers of porous sheet material in the humidification portion which cover and support substantially the entire surface of the membrane. Similarly, the grooving and sealing techniques employed on the porous electrically conductive sheet material in the electrochemically active portion can also be advantageously employed on the porous sheet material in the humidification portion of a fuel cell stack.

Many further modifications will readily occur to those skilled in the art to which the invention relates and the specific embodiments described herein should be taken as illustrative of the invention only and not as limiting its scope in accordance with the accompanying claims.

Claims

1. A membrane electrode assembly for an electrochemical fuel cell comprising first and second layers of porous electrically conductive sheet material and a membrane interposed

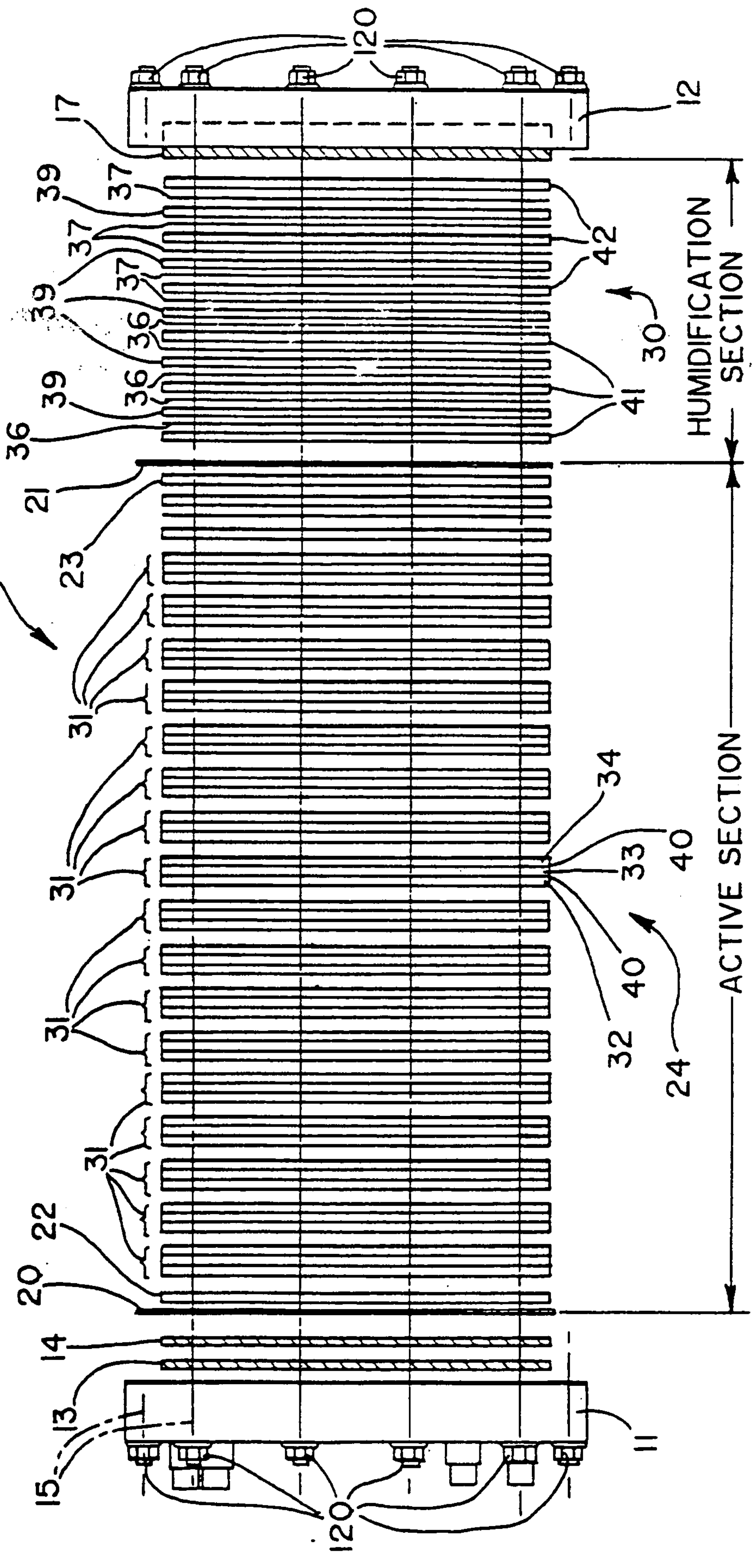

120
12
17
37
39
42
30
HUMIDIFICATION SECTION
36
41
21
23
10
31
34
33
40
32
24
ACTIVE SECTION
22
20
14
13
11
15

Fig. 1

Fig. 5

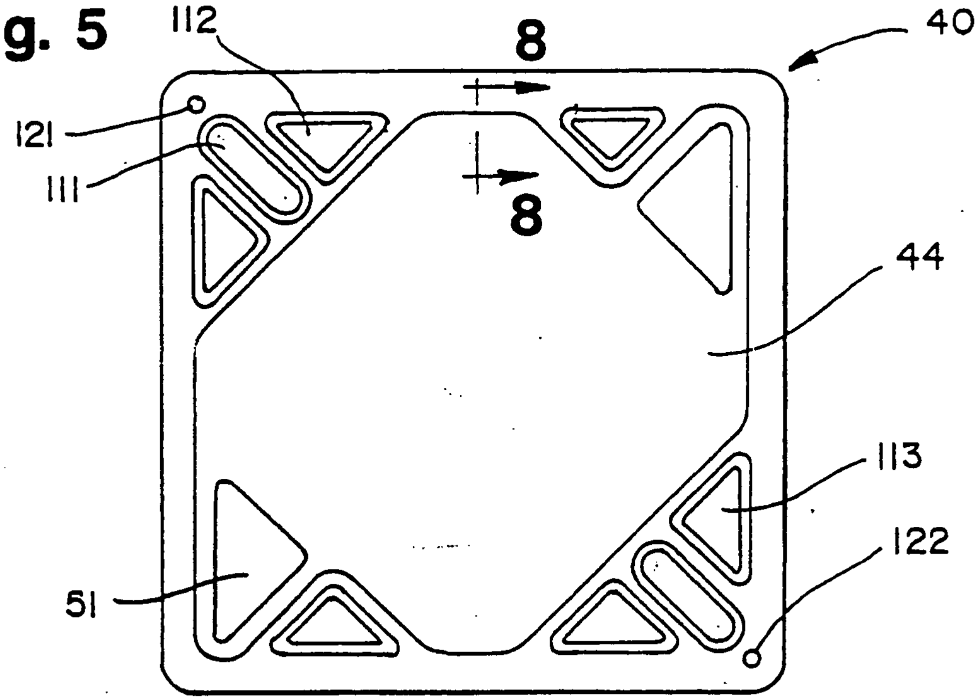


Fig. 6

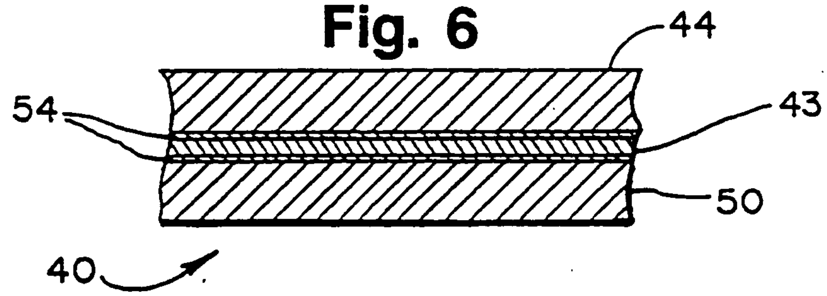


Fig. 7

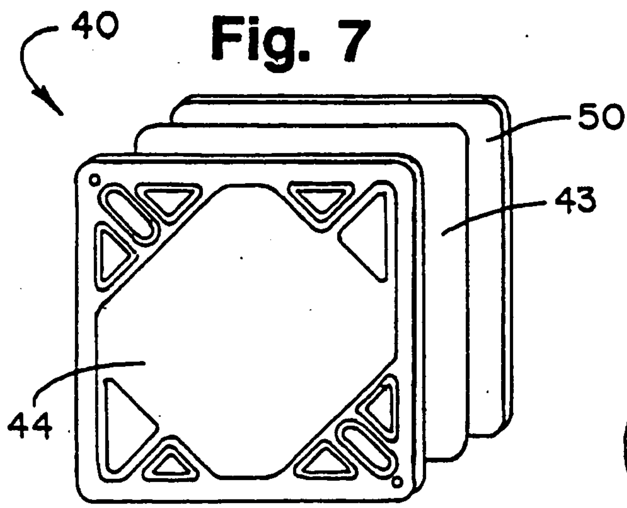
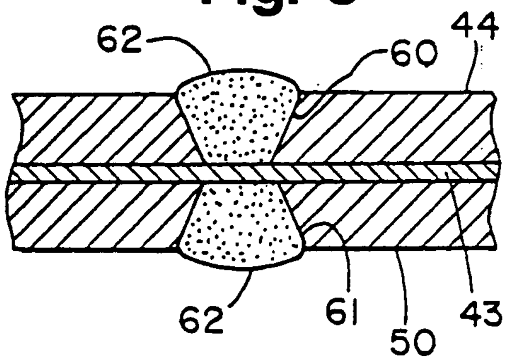


Fig. 8



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EUROPEAN SEARCH REPORT

Application Number

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DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A	PROCEEDINGS OF THE 26TH INTERSOCIETY ENERGY CONVERSION ENGINEERING CONFERENCE - IECEC-91 vol. 3, 4 August 1991, MASSACHUSETTS pages 636 - 641 M. S WILSON ET AL 'RECENT ACHIEVEMENTS IN POLYMER ELECTROLYTE FUEL CELL (PEFC) RESEARCH AT LOS ALAMOS NATIONAL LABORATORY' * page 638, left column, paragraph 2 *	1-7,9	
A	US-A-3 012 086 (NATHAN P. VAHLDIECK) * claim 1; figure 1 *	10	
E	WO-A-9 313 566 (INTERNATIONAL FUEL CELLS INC.) * page 12, line 28 - line 34; figure 2 * * page 14, line 19 - line 32; claim 1 * * page 9, line 19 - page 10, line 14 *	1-2,5,10	
E	US-A-5 176 966 (DANNY G. EPP ET AL) * the whole document *	1-10	TECHNICAL FIELDS SEARCHED (Int. Cl.5)

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	07 SEPTEMBER 1993	D'HONDT J.W.

CATEGORY OF CITED DOCUMENTS

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